Microstructure evolution of high capacity anode electrode by in-situ and in-operando X-ray nano-CT

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Alloy-typed materials have been studied as an anode active material to develop high energy density lithium ion batteries (LIBs). Especially, lithium alloys based on the group IV elements (Si, Ge, and Sn) are potential candidates for the anode material because of their high theoretical capacities and low operating voltages. Lithiation and delitiation of the anode alloys accompany large volume change that causes fractures, pulverizations, and delamination of the electrodes. The mechanical degradation reduces the reversible capacity and shortens the cycle life of the alloy anode LIBs. Particle fracture has been alleviated by nano-structuring the alloy-type anode materials due to the facile strain accommodation and the short diffusion path for electron and lithium ion transport in these nanostructured electrodes. However, nano-structured particles have low tap density and lead to lower energy density anodes, making scale up difficult. The surface area of the material increases with decreasing particle size, which leads to large irreversible capacity loss due to the formation of the solid electrolyte interphase (SEI). Currently, a fundamental understanding of the impact of a high capacity electrode's microstructure change on LIB performance is still lacking due to the inhomogeneity, complexity, and 3D nature of the electrode's microstructure.

In this study, a novel approach is proposed to gain greater understanding of the microstructure change of the alloy anode electrodes and its impact on the electrochemical performance. A special LIB cell was designed to monitor the microstructure change of high capacity anode electrodes with the synchrotron X-ray nano-CT technique at the Advanced Phothon Source of Argonne National Lab. The cell is composed of a quartz capillary housing and a wire-typed electrode to maximize X-ray penetration for the nano-CT scan. The structural evolution of the alloy electrodes is monitored to investigate crack propagations and pulverizations under in-operando 2D x-ray CT scan. Moreover, in-situ 3D x-ray CT scan enables to study the anisotropic volumetric changes at different voltage states. This simultaneous structural and electrochemical investigation of the alloy electrodes is an essential study to understand the fundamental degradation mechanism of high capacity lithium alloy anode.